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PARTICLE TRACKING NUMERICAL METHODS FOR NANOPARTICLE TRANSPORT IN HETEROGENEOUS POROUS MEDIA

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Abstract. A single-phase flow, lattice Boltzmann method (LBM) is utilized with a Lagrangian particle tracking (LPT) method for the simulation of flow and transport of nanoparticles in a porous medium. The 3D pore matrix is obtained either as a randomly packed with spheres porous medium or from images of segments of rock (sandstone) through micro-computed tomography (μ-CT). The particles are assumed to be passive. When the particles collide with the solid matrix, they can either adsorb or continue their motion, based on the assumption that the deposition process is a pseudo-first order process. Furthermore, the solid-fluid interface is assumed to be heterogeneous, so that the simulated nanoparticles can adsorb at different rates at different sites of the interface. Simulations are validated with theoretically expected results, based on macroscopic filtration equations.

1 INTRODUCTION

Charge heterogeneity of the pore surfaces has been found to be important for colloidal transport in porous media. In such conditions, the presence of surface charge heterogeneity generates patchy surface areas, which can be favourable for particle attachment [1, 2]. It is widely admitted that the surface charge heterogeneity in natural porous media, like rocks in hydrocarbon reservoirs, originates from metallic oxides (such as iron, aluminum, and manganese oxide) being present at the porous matrix surface. Naturally, these oxides are randomly distributed across the porous medium (often called the collector) surfaces, yet they are usually grouped into patches in mathematical models. This type of modeling results in the assumption of a patchy distribution of surface charge heterogeneity [3]. It has been claimed so far in the literature that the pattern of the patchy distributions plays no role in colloidal retention, yet the fraction of favorable surface, λ, caused by heterogeneity is important [4].

In this work, we use lattice Boltzmann methods in conjunction with Lagrangian particle tracking (LBM/LPT) simulations to numerically study the effect of different patterns of surface charge heterogeneity in packed-sphere beds. We created four different patterns of surface charge heterogeneity, and conduct parametric investigation of λ and probability of attachment associated with the heterogeneous surfaces under different pore velocities.

2 LATTICE BOLTZMANN METHOD AND LAGRANGIAN PARTICLE TRACKING

To simulate the velocity field in the packed-sphere beds, we adopted the lattice Boltzmann method (LBM), a class of computational fluid dynamics methods with second-order accuracy. This method has been widely adopted to simulate flow of single phase and multi-phases over a wide range of Reynolds number flows. Details about the method can be found elsewhere [5, 6], and are not presented herein. The use of the LBM begins with choosing the number of directions in space and the number of velocity lattice vectors, typically denoted as *DmQn*, where *m* is the former and *n* is the later. We employ a D3Q15 scheme with a bounce back boundary condition in this study. The packed-sphere beds were numerically generated by packing rigid and impermeable spheres in simulation boxes following a modified Lubachevsky-Stillinger algorithm [7]. Water with a viscosity of 0.001 Pa.s was used as the working fluid.

Table 1. Simulation parameters

Sphere diameter, d_S	250μm
Porosity, ϕ	*0.46*
Pore velocity, u_p	*0.02, 0.065, and 0.13 cm/s*
Fraction of attachable surface ($p_a \neq 0$), λ	0.2, 0.5, and 0.8
Deposition probability, p_a	0.001, 0.005, 0.01, and 0.05
Number of injected pore volume, PV	5
Particle diameter, d_p	10 and 53 nm

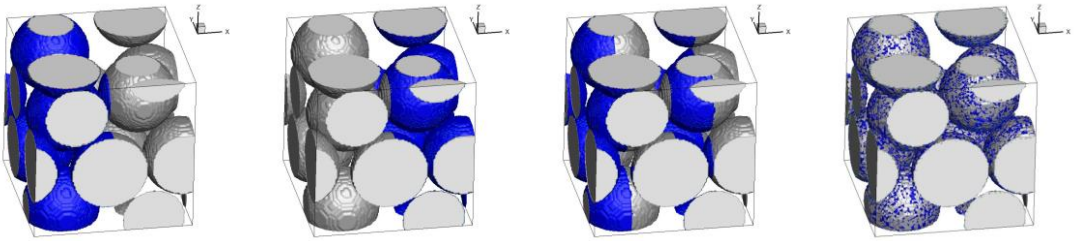

Figure 1. Different spatial distributions of surface charge heterogeneity. Assuming flow from left to right, the blue surface areas indicate surfaces where deposition can occur (i.e., $p_a \neq 0$), whereas those in gray are non-depositable ($p_a = 0$). From left to right: *Entry*, *Exit*, *Stripes*, and *Mixture*.

In associated with the LBM, the Lagrangian particle tracking (LPT) algorithm is employed to advance passive particles through the packed-sphere beds. This method allows one to follow the particles and track their trajectories in the Lagrangian framework. As long as dilute particle solutions are considered in this study, the particles are assumed to be monodisperse with no particle-particle interactions. At each simulation time step, the particles are propagated by convection and Brownian motion. When colliding with the solid wall, the particles can either be returned to their original positions at the previous time step or attached to the wall with a pre-defined probability of attachment, p_a. A particle is retained if the pre-defined p_a is larger than or equal to a random number between 0 and 1, generated at the time of collision. Notice that irreversible attachment is assumed in the attachment implementation. Details of the mathematical formulation of the LPT is available in our previous works [8-10].

In this study, we generated four different packed-sphere beds with four different patterns of surface charge heterogeneity, named Entry, Exit, Stripes, and Mixture (see Figure 1). To simulate the heterogeneous surface charge, p_a was non-zero at λ fraction of the surface area, and was zero otherwise. The particle injection strategy consisted of 5 pore volume (PV) of passive particles in each simulation. Other simulation parameters are available in Table 1.

3 RESULTS AND DISCUSSION

3.1 Validation

The precision of the LBM/LPT in conjunction with the probabilistic approach of particle deposition checking in the presence of chemically heterogeneous collector is validated by matching simulation results with that predicted from patch-wise model. Due to irreversible attachment, the patch-wise model is formulated as follows

$$\begin{cases} \dfrac{\partial C}{\partial t} = D\dfrac{\partial^2 C}{\partial x^2} - u\dfrac{\partial C}{\partial x} - \dfrac{\rho_b}{\phi}\dfrac{\partial S}{\partial t} \\ \dfrac{\partial S}{\partial t} = \dfrac{\phi}{\rho_b} k_a C, \qquad k_a = \lambda k_{a-high} + (1-\lambda) k_{a-low} \end{cases} \tag{1}$$

where C is the concentration of particles, D is the effective diffusivity of the particles in the porous media, u is the pore velocity, ρ_b is the bulk density of the column, ϕ is the porosity of the column, S is the adsorbed particle concentration, k_a is the average attachment coefficient. Figure 2 presents breakthrough data, obtained from simulation along with prediction from Equation (1). Simulation conditions can be found in the figure caption. It is evidenced from Figure 2 that the simulation results are in excellent agreement with the prediction, confirming the precision of the simulation approach.

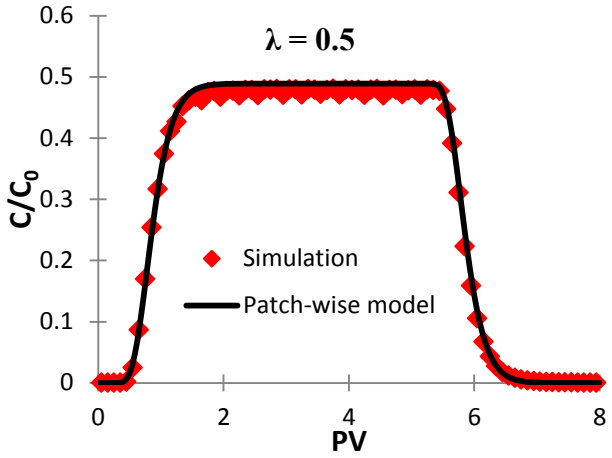

Figure 2. Comparison of particle breakthrough curves obtained from simulation and patch-wise model. Simulations was done with d_S = 150 μm, d_p = 100 nm, ϕ = 0.35, u_p = 0.0028 cm/s, $k_{a\text{-high}}$ = 0.105 1/s, $k_{a\text{-low}}$ = 0.01 1/s, and D = 7×10^{-6} cm^2/s.

3.2 Effect of probability of attachment, p_a

To look into the effect of p_a on particle retention, we isolate the contribution of λ by fixing its value at 0.5 while pore velocity was varied from 0.02 to 0.13 cm/s. Also, p_a took four different values of 0.001, 0.005, 0.01, and 0.05. In our simulation, p_a is proportional to the favorable level of the collector surface, and is correlated to the nominal deposition rate as follows

$$k_0 = \frac{1}{\Delta t}\ln\left(\frac{1}{1-p_a}\right) \tag{2}$$

where Δt is the simulation time step. Figure 3 shows the average attachment coefficient, ka, as a function of Da×Pe. Note that ka was obtained by fitting simulation data with Equation (1), and Da is the Damkohler number and Pe is the Peclet number, which are dimensionless. It is apparent from Figure 3 that the pattern effect is noticeable at Da×Pe equal to 400 and beyond, as ka of the four different heterogeneous patterns start going apart. The difference between ka is more pronounced at higher Da×Pe, inferring the role of the heterogeneous pattern at high pa. The fact that the divergence of k_a is only visible at a certain Da×Pe, a threshold of Da×Pe beyond which the pattern effect is notable might exist. Among the four heterogeneous pattern, beyond the diverging point, k_a of the *mixture* is always the highest.

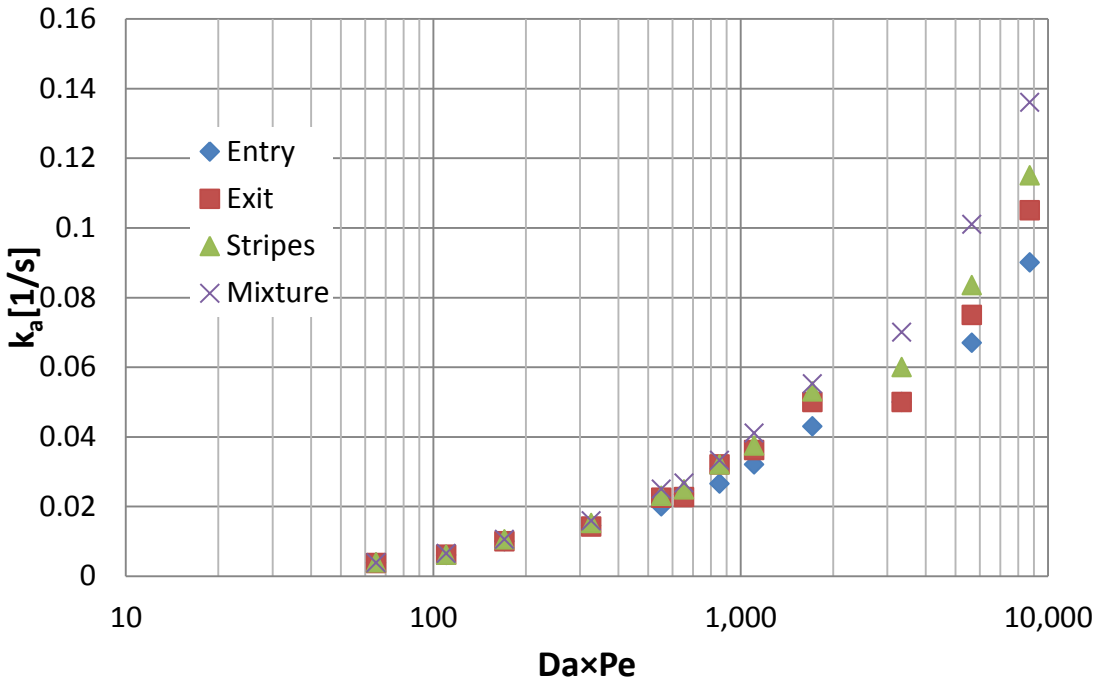

Figure 3. Particle deposition rate as a function of the product of Damkohler and Peclet number at different p_a and pore velocities. Other simulation parameters included d_p = 10 nm and $\lambda = 0.5$. p_a were 0.001, 0.005, 0.01, and 0.05, u_p was varied at *0.02, 0.065, and 0.13 cm/s.*

12 CONCLUSIONS

- Our findings have shown that the effect of surface charge heterogeneity pattern play an important role in colloidal retention in packed beds. At high deposition rate, the effect becomes more pronounced.
- On the other hand, it diminishes as the deposition rate decays.
- At a particular λ, there exists a threshold of Da×Pe beyond which the pattern effect should be taken under consideration.

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